

# Mobile Anti-phase Domains in Lightly Doped Lanthanum Cuprate

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Light hole doping of lanthanum cuprate strongly suppresses the onset of antiferromagnetic (AF) order. Surprisingly, it simultaneously suppresses the extrapolated zero temperature sub-lattice magnetization.  $^{139}\text{La}$  NQR results in lightly doped  $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$  have demonstrated that these effects are independent of the details of the mobility of the added holes. We propose a model in which doped holes phase separate into charged domain walls that surround “anti-phase” domains. These domains are mobile down to  $\sim 30\text{ K}$  where they either become pinned to the lattice or evaporate as their constituent holes become pinned to dopant impurities.

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## I. INTRODUCTION

A fundamental issue in the normal state of the superconducting cuprates is the behavior of holes doped into a two-dimensional lattice of spins with strong antiferromagnetic (AF) interactions. Even for lightly doped, single layer lanthanum cuprate many important issues remain poorly understood. Long-range antiferromagnetic order occurs at  $T_N > 300\text{ K}$  in undoped lanthanum cuprate, but  $T_N$  is rapidly suppressed by the addition of a small density,  $p$  of holes per Cu. This rapid suppression is clearly related to the disruptive effects of mobile holes:  $p \lesssim 3\%$  is sufficient to suppress  $T_N$  to zero, while  $\sim 30\%$  isovalent substitution of Zn or Mg for Cu is required [1] to produce the same effect. A range of studies [2] including  $^{139}\text{La}$  NQR measurements [3] in lightly doped  $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$  have demonstrated that the suppression of  $T_N$ , and in fact, all the magnetic properties of lightly doped lanthanum cuprate are essentially *invariant* without regard for the means of hole doping and consequent variations in hole mobility.

It is unlikely that a collection of individual holes can lead to magnetic behavior that is entirely independent of compositional variation that leads to substantial variations in resistivity (at constant doping). We argue, instead, that this is strong evidence that holes form collective structures. An important and well documented aspect of doped cuprates is their tendency toward inhomogeneous charge distribution [4]. Segregation of doped holes into charged stripes separating hole-free domains has been predicted [5–11] and recently observed directly in lanthanum cuprate [12]. It was proposed earlier that phase segregation of holes could be responsible for the unusual magnetic properties of lightly Sr-doped lanthanum cuprate [13–15]. We make a related proposal that holes form charged, domain walls which form closed loops with the important differences that these walls form anti-phase domain walls (so the the phase of the AF order inside

these domains is reversed) and that the walls and hence the enclosed domains are mobile, and the charged walls have the density of 1 hole per 2 Cu sites in agreement with neutron scattering results [12]. The anti-phase character means that mobile (above 30 K) domains will suppress the time-averaged static moment thus suppressing  $M_s$  as well as  $T_N$ . These domain structures will have contrasting interactions with in-plane *vs.* out-of-plane dopants (*e.g.*, stronger scattering by in-plane impurities) which explain the different transport behaviors, while the universal magnetic properties can be understood as long as the domains are sufficiently mobile that they move across a given site rapidly compared to a measurement time.

## II. LIGHTLY DOPED LANTHANUM CUPRATE

A systematic study of the temperature  $T$  and doping dependence of the static susceptibility in lightly doped lanthanum cuprate by Cho *et al.* [13] provided evidence that the added holes are inhomogeneously distributed. The development of long-range antiferromagnetic order is signaled by a peak in the static susceptibility; they showed the rapid increase in the width of this peak with increasing hole density could be understood as arising from finite-size effects. They proposed that doped holes form hole-rich domain walls that bound hole-free domains, thus cutting off spin interactions across the boundary and truncating the growth of the spin-spin correlation length with decreasing temperature above  $T_N$ . They deduced the doping dependence of the dimension  $\mathcal{L}$  of the hole free-regions, and found  $\mathcal{L} \simeq (0.02/p)^2$ ; this suggests that the density of holes within the boundary stripe is very low,  $\sim 1$  hole per 5 or 10 Cu sites.

$^{139}\text{La}$  NQR measurements in lightly Sr-doped lanthanum cuprate by Chou *et al.* provided a detailed picture of the  $T$  and  $p$ -dependence of its magnetic properties [14]. They found that for  $30\text{ K} \lesssim T < T_N$  the sublattice

FIG. 2. The relationship between  $M_s^0(p)$  and of  $T_N(p)$  as both are suppressed by increasing doping  $p$ .  $M_s^0(p)$  is obtained as explained in the text from  $^{139}\text{La}$  NQR data for  $\Delta\nu$  such as is shown for the Li case in Fig. 1. The closed circles are from  $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$  [3], and the open circles are from  $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$  [15]. The results are the same in both materials, illustrating one aspect of the similarity of the magnetic properties, in spite of the much higher resistivity found in  $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$  as a consequence of the in-plane impurities. The solid line is due to Castro Neto and Hone [16].

to the Sr doping case [3]. Comparing  $\text{La}_{2-y}\text{Sr}_y\text{CuO}_4$  (LSCO) and  $\text{La}_2\text{Cu}_{1-x}\text{Li}_x\text{O}_4$  (LCLO) at  $x = y = p = 0.025$  one finds that the room temperature resistivity of LCLO [2,17] exceeds that of LSCO [18] by over an order of magnitude. Furthermore, unlike LSCO, the resistivity of LCLO always increases monotonically with decreasing temperature. With increasing doping the contrast becomes more dramatic as LSCO becomes metallic and superconducting while LCLO becomes ever more insulating with doping above  $p = 0.1$ .

In spite of this we find that the magnetic behavior of the two materials is essentially identical [3]. In addition to the similarly strong suppression of  $T_N$  by doping [2], we find that  $M_s$  is also suppressed, and the correspondence between the suppression of  $M_s$  and  $T_N$  by doping is identical to that observed in LSCO [15]. In Fig. 2  $M_x^0(p)/M_x^0(0)$  for both LCLO (Ref. [3]) and LSCO (Ref. [15]) is plotted against  $T_N(p)/T_N(0)$ . Here  $M_s^0$  is the value of  $M_s$  obtained by extrapolating the  $M_s(T)$  data for  $T > 30\text{ K}$  to  $T = 0$  i.e., the  $T = 0$  value of the solid lines shown in Fig. 1(a). The solid line through the data is due to a theory of Castro Neto and Hone [16]; see also van Duin and Zaanen [19]. The strong peak in  $2W$  occurs at the same temperature and exhibits the same binding energy (as extracted from the  $T$ -dependence on the high temperature side of the peak) [3]. Finally, the temperature dependence of the low-energy dynamical susceptibility (obtained from measurements of  $2W(T)$ ) exhibits the same finite-size effects [3] as were observed in the static susceptibility by Cho *et al* [13].

FIG. 4. Upper panel: The variation of the size  $l$  of the anti-phase domains required to explain the observed suppression of  $M_s^0$  by doping is shown along with the average size  $L$  of the region which encompasses a single anti-phase domain. The inset to the lower panel shows the data of Borsa *et al.* for  $R(p) \equiv M_s^0(p)/M_s^0(0)$  [15] along with the parametrization of the  $p$ -dependence used to calculate the lengths shown here;  $p_0 = 0.028$ . Lower panel: The variation of  $\mathcal{L}^2$  with doping is plotted against the left-hand axis, and the results of Cho *et al.* [13] are plotted against the right-hand axis. The fit obtained by scaling the single parameter which sets the overall magnitude of  $f(p)$  obtained by Cho *et al.* is very good. Also shown, plotted against the left-hand axis, is the fit  $(0.4/x)^2$  suggested by Cho *et al.* above  $p = 0.01$ .

$$R(p) = 1 - (p/p_0)^2 \quad (1)$$

with  $p_0 = 0.028$ . For simplicity we assume that a (1,0) or (0,1) domain wall orientation is preferred, and so consider square domains.

If a region of size  $L$  contains, on average, one anti-phase domain of size  $l$  (see Fig. 3, all lengths are in units of the lattice parameter), then

$$R = 1 - (2N_-/N) \quad (2)$$

Here  $N = N_+ + N_- = L^2$ , where  $N_- = l^2$  is the number of sites in the anti-phase domain and  $N_+$  is the number of sites in the dominant AF phase. The number of holes in the region of size  $L$  is  $pL^2$ ; the domain wall which bounds the anti-phase domain contains 1 hole per 2 Cu sites, so  $4l = 2pL^2$ . From Eqns. 1 and 2

$$L^2 = N = 2(1 - R)/p^2 = 2/p_0^2 \quad (3)$$

and

$$l = (1 - R)/p = p/p_0^2 \quad (4)$$

hence  $L \simeq 50$ . The variation of  $l$  with  $p$  based on the experimentally determined variation of  $R(p)$  is shown in Fig. 4(a). It should be noted that the behavior found here is particularly simple as a consequence of the parametrization of  $R(p)$  chosen (Eq. 1); this parametrization is not uniquely determined by the data.

This simple model has several appealing features. The model described in Section II which relies on static domain walls implies a very low hole density in the wall ( $\sim 0.1$ – $0.2$  holes/Cu site) which must nonetheless maintain its integrity as a charged stripe and entirely cut off AF interactions across the stripe. Our model posits a density of 0.5 holes per Cu site such as is observed in neutron scattering and predicted by calculations [29]. The recovery of  $M_s$  below 30 K is straightforwardly understandable since once motion of the anti-phase domains becomes slow compared to the NQR time scale ( $\sim 0.1$ – $1$   $\mu$ sec) time averaging of the reversed spin directions will cease and the full ordered moment will be observed. This could arise either from pinning of the anti-phase domain to the lattice or evaporation of the domain walls due pinning of the constituent holes to the charged donor impurities; in either case the coincidence of the recovery of  $M_s$  and the freezing of spin degrees of freedom evidenced by the low  $T$  peak in  $2W$  is naturally explained. The correspondence between suppression of  $M_s^0$  and  $T_N$  is natural in this case because interlayer coupling will be hampered wherever an anti-phase domain is present, thus impeding the development three-dimensional AF ordering. See the discussion in Ref. [29] in this regard.

This model also explains the finite-size effects revealed by the susceptibility analysis of Cho *et al.* [13] if we consider that the appropriate length scale between domain walls is  $\mathcal{L} = (L-l)$ . The variation of  $\mathcal{L}^2$  with  $p$  is shown in Fig. 4(b) and compared with the variation of the square

of the characteristic length scale obtained by Cho *et al.* [13] (scaled vertically to obtain the best agreement). Finally we note from Fig. 4(a), that  $L$  and  $l$  converge with increasing  $p$ , and we expect that loops will cease to be stable when  $L$  approaches  $l$ . For the parametrization of  $R(p)$  we have chosen,  $L = l$  when  $p = \sqrt{2}p_0 = 0.04$ , near the doping at which the the metal-insulator transition and spin-glass behavior are found. We speculate, then, that these are related to the transition in the configuration of the charged domain walls from loops to parallel stripes.

In conclusion, we have presented a model which explains the range of unusual magnetic phenomena observed in lightly doped lanthanum cuprate. In particular, we can understand the insensitivity of magnetic properties to materials variations that substantially increase the resistivity. This indicates that mobile anti-phase domains play a central role in determining the magnetic properties of lightly doped lanthanum cuprate. It may point to an explanation of the poorly understood “spin-glass” regime of the phase diagram in terms of a crossover in domain wall topology from loops to parallel stripes. More generally, it suggests that the development of stripe order may play a determining role in the phase diagram of the cuprates (see e.g., Ref. [30]). Rather than requiring mobile domain walls, superconductivity may more sensitively depend on the nature of the ordering of the walls into parallel stripes.

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